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Assessing the Influence of Different Atmospheric and Soil Mercury Concentrations on Foliar Mercury Concentrations in a Controlled Environment

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Abstract This study investigated the influence of soil and air mercury (Hg) concentrations on Hg accumulation in plant components for three plant species, *Robinia pseudoacacia* (Black locust), *Juniperous scopulorum* (Juniper), and *Artemisia tridentata* (Sagebrush), grown in environmentally controlled growth chambers. Exposures included ambient and elevated air (3.1±0.4 and 30.1±3.5 ng m^{-3}) and soil (0.06±0.02 and 27.7±6.3 μg g^{-1}) Hg concentrations. In addition, foliar Hg fluxes were measured, as was Hg accumulated directly on the leaf surface. Air Hg concentrations were found to be the dominant factor associated with foliar Hg concentrations. Foliar MeHg concentrations of deciduous plant species were greater than evergreen species. Trunk Hg concentrations were influenced by air and soil Hg concentrations. Root Hg concentrations were directly correlated with soil Hg concentrations. Foliar Hg fluxes for *R. pseudoacacia* were predominantly deposition. For *A. tridentata* foliar Hg fluxes were bi-directional, and foliar fluxes measured for *J. scopulorum* were not statistically different from the blank chamber fluxes. Measured fluxes did not correspond well with predicted uptake based on foliar Hg concentrations.

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1 Introduction

Global ambient atmospheric mercury (Hg) concentrations are ~1.6 ng m^{-3} (Lamborg, Fitzgerald, O'Donnell, & Jorgensen, 2002; Lindqvist et al., 1991; Schroeder & Munthe, 1998) and in mining areas may be as high as 200 ng m^{-3} close to the ground (Gustin et al., 1999). Mercury exists in the atmosphere as predominantly gaseous elemental mercury (Hg^o) (>95%) (Fitzgerald, Mason, & Vandal, 1991) with lesser amounts of Hg(II) containing species, collectively designated as reactive gaseous mercury (RGM: $HgBr_2$, $HgCl_2$, $Hg(CH_3)Cl$), and particulate bound forms (Lindqvist et al., 1991). Methyl mercury (MeHg) has been suggested to be ≤1% of the total mercury (THg) in the atmosphere (Schroeder & Munthe, 1998).

Natural sources of Hg to the atmosphere include enriched and unenriched soil, fresh and salt-water bodies, vegetation, wildfires, out gassing of the earth's crust, volcanoes and geothermal sources (Nriagu, 1989; Schroeder & Munthe, 1998). Mercury is also emitted by anthropogenic sources including ore processing facilities, fossil fuel combustion, and sewage sludge and medical waste incineration (Schroeder & Munthe, 1998).

Atmospheric Hg may be deposited to soil and plant surfaces by wet and dry processes (Schroeder &

Munthe, 1998). Deposited Hg may be subsequently reemitted from plants and soil surfaces back to the atmosphere (Ericksen, Gustin, Lindberg, Olund, & Krabbenhoft, 2005; Hintelmann et al., 2002). A dominant factor found to be correlated with foliar Hg concentrations is air Hg concentration (Ericksen & Gustin, 2004; Millhollen, Gustin, & Obrist, 2006), while soil Hg concentrations have been to shown to be less well correlated (Ericksen et al., 2003; Fleck, Grigal, & Nater, 1999; Frescholtz, Gustin, Schorran, & Fernandez, 2003; Millhollen et al., 2006). In addition plants have been demonstrated to transport Hg from the soil through the plant and this Hg may then be emitted to the atmosphere (Hanson, Lindberg, Tabberer, Owens, & Kim, 1995; Leonard, Taylor, Gustin, & Fernandez, 1998). Bishop, Lee, Munthe, and Dambrine (1998) reported up to 11% Hg transport and up to 3% MeHg transport from the soil into plant tissue based on measurements of Hg in the xylem sap of *P. abies* and *P. sylvestris*, but did not measure Hg emission to the atmosphere. Greger, Wang, and Neuschutz (2005) found that terrestrial plants (*P. sativum, T. aestivum, B. vulgaris, B. napus, T. repens, S. viminalis x schwerinii*) grown in elevated Hg soil translocated 0.17–0.25% of Hg from the sediment to the shoot of the plant, and they reported no release of Hg to the atmosphere.

In this study three plant species *R. pseudoacacia*, *J. scopulorum*, *A. tridentata* were grown in soils with natural background Hg concentrations (≤0.1 μg g^{-1}) (Craig, 1986; Ulrich, Tanton, & Abdrashitova, 2001) and high Hg concentrations (30 μg g^{-1}) and at ambient and elevated air Hg exposures. Foliar Hg fluxes were measured for all three species using a single pass gas exchange chamber. Leaf samples were collected in five harvests over the growing season to assess foliar Hg concentrations. With each harvest leaf washes were collected to determine dry Hg deposition to the leaf surfaces. At the end of the experiment foliar root and trunk Hg concentrations were measured. This study was designed to provide data that could be compared to that collected in a previous study by Millhollen et al. (2006) who applied a similar experimental design but utilized different forms of Hg in the soil. One of the goals was to see if Hg speciation in the soil had any influence on plant total Hg accumulation or flux. This study also quantified MeHg concentrations in foliar tissue from *R. pseudoacacia* and *J. scopulorum* to see if there were any differences in concentration as a function of plant species. Tissues used for MeHg analyses were obtained from this study and from the experiments done by Millhollen et al. (2006).

2 Materials and Methods

2.1 Experimental design

Three plant species, *R. pseudoacacia* (Black locust), *J. scopulorum* (Juniper), and *A. tridentata* (Sagebrush) purchased in November as 12-month-old saplings (Nevada Division of Forestry: Washoe Nursery, Carson City, NV, USA) were utilized in this experiment. The saplings were grown in an outdoor nursery and all saplings had foliage on the date of purchase. Saplings were repotted in two soil types; topsoil (0.06±0.02 μg g^{-1}) purchased from Rainbow Rock, Reno, NV, and topsoil amended at a 1:10 ratio with elevated Hg containing substrate collected from the property of Ormat Geothermal, Reno, NV in the Steamboat Springs Geothermal Area. This area is naturally Hg-enriched with cinnabar and elemental Hg (Coolbaugh, Gustin, & Rytuba, 2002), and the substrate was collected from Hg mine tailings and had a concentration of ~176 μg g^{-1}. The topsoil was homogenized with the naturally enriched substrate using a cement mixer to achieve a soil Hg concentration of 27.7±6.3 μg g^{-1}. At the end of the experiment Hg in the soil column of select pots was measured to determine if Hg had been redistributed within the soil column. The Nevada Bureau of Mines and Geology analyzed the substrate for Hg by atomic absorption spectroscopy after digestion in aqua regia (Lechler, 1999).

Plants were grown in four environmentally controlled, closed-system growth chambers, or Ecopods, located in a naturally lit greenhouse (Fritz Went Laboratory, Desert Research Institute, Reno, NV) (cf. Frescholtz et al., 2003). The chambers have dimensions of 1.4 m×1.0 m×1.5 m with all sides consisting of glass walls. Twenty plants of each species were grown in the four Ecopods with 10 of each plant species in background Hg concentration soil and ten of each species in Hg enriched soil (Fig. 1).

The air in two Ecopods was maintained at elevated atmospheric Hg concentrations of 25–30 ng m^{-3}

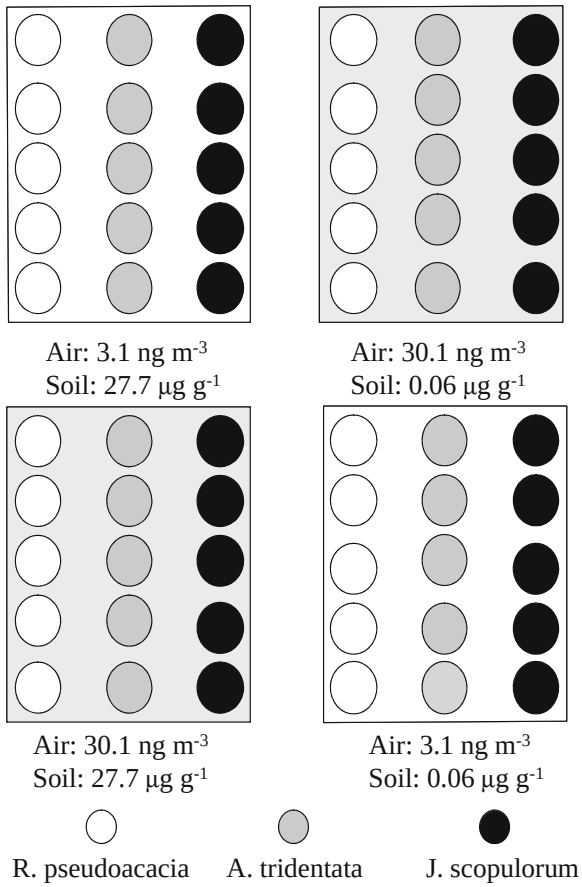

Fig. 1 Experimental design for each Ecopod including plant species and air and soil Hg exposure concentrations

(30.1±3.5 ng m^{-3}), and two had ambient Hg concentrations of 2–5 ng m^{-3} (3.1±0.4 ng m^{-3}). Gaseous mercury was produced using elemental Hg permeation tubes (VICI Metronics, Poulsbo, Washington, USA) that were contained in glass housing and submerged in a constant-temperature (38°C) water bath. Flow through the permeation source housing headspace was controlled to regulate the release of Hg° into the Ecopods. Mercury concentrations were monitored continuously over the experiment using a mercury analyzer (Tekran® 2537A Toronto, Canada) attached to a solenoid-switching unit that allowed for sequential sampling of the air in each Ecopod.

Plant pots were made of polyvinyl chloride pipe (0.15 m (diameter), 0.44 m (length)) that were sealed at the bottom with a pipe cap. The top of the pipe was covered with two clear Plexiglas® pieces that were sealed with silicon caulking. The center of the Plexiglas® top had a hole for the plant stem and was sealed with polyethylene foam, and covered with Polyfin™ wax. Each Plexiglas® lid had an inlet and an outlet port to allow for headspace air (~1.5×10^{-3} m^2) to be continuously recharged with ambient air from outside the greenhouse. A third port, closed with a silicon cork, was used for watering.

Weekly pots were weighted and watered, and plants visually inspected for insect infestation, senesced foliage, and damage. Greenhouse tap water was used to water the plants throughout the experiment and Hg concentrations in the water were measured monthly (1.91±0.73 ng L^{-1}). Based on the initial dry soil weight, soil moisture was maintained at 8–12% and no water drained from the pots. After planting and prior to the soil being sealed off, each pot was fertilized with 15 Osmocote® pellets on the soil surface.

Two days after planting, before sealing and placement in the Ecopods, foliar tissue samples were collected to represent an initial base line Hg concentration (Harvest 1). All plants were then placed in chilled (9.7±0.86°C day, 8.4±0.36°C night) Ecopods (n=30 days, November–December) to simulate winter conditions. The plants remained dormant for 1 month in the chilled Ecopods and at the end of the month, foliar tissue samples were collected (Harvest 2, t=4 weeks) from *J. scopulorum* and *A. tridentata,* but not *R. pseudoacacia* because all foliage senesced due to the cold temperatures. At the end of December, air temperature within the Ecopods (December–August) was increased and maintained at 25.1±2.0°C during the daytime (06:30–17:30) and 17.7±0.2°C at nighttime (17:30–06:30).

Foliar tissues that had been on the plant for >1 month were collected every four weeks using clean handling procedures (cf. Frescholtz et al., 2003). Leaves were rinsed in ultra pure de-ionized water in acid cleaned Petri® dishes, and water samples from each harvest were preserved in 100 ml flasks with 1% Optima HCl until analyzed. For a subset of leaves for each species leaf area was measured with a Portable Area Meter (LI-COR model LI-3000A) along with corresponding leaf length, width and dry weight. These measurements were used to develop allometric relationships for determination of leaf area (Hicks & Dugas, 1998; Svenson & Davies, 1992). All allometric relationships had $r^2 \geq 0.80$ and $p<0.05$ (Fay, 2006).

During the experiment gnats were present on *R. pseudoacacia* in all treatments and caused minor leaf

deformation. *R. pseudoacacia* in the ambient air-low soil treatment were treated with spray insecticide (Dr. Earth, Organic Pro-active Insect Spray) once prior to Harvest 3 to test its effectiveness. The insecticide had Hg concentration of 25.0±5.0 ng L^{-1}, with 0.58 ng m^{-2} of Hg applied. The applied insecticide was rinsed off the *R. pseudoacacia*. Gnats were subsequently removed by hand with Kimwipes® prior to each watering. Because subsequent leaf wash concentrations were low (1.5±1.1 ng m^{-2}) and were not statistically different from those collected from leaves that did not have an insecticide application, it was assumed that all the insecticide was washed off.

At the end of the experiment all plants were separated into the following components: foliar tissue, trunks, and roots (for only *J. scopulorum* and *R. pseudoacacia*). An aliquot of each sample was taken for Hg analyses, and the remaining tissue was dried in an oven for 48 h (60°C) to allow for calculation of the total Hg in each plant part. *J. scopulorum* was harvested at week 17, and *R. pseudoacacia* and *A. tridentata* were harvested at week 21.

2.2 Laboratory analyses

After sampling, harvested plant tissue was homogenized, lyophilized for 24 to 48 h using a Virtis Benchtop 3L (Sentry™), and stored at −20°C until analyzed. Total Hg content in the plants was determined using a Milestone™ Direct Mercury Analyzer (Model DMA 80, AMA 254 Software) (EPA method 7473). Analyses of standard reference material (NIST 1547 peach leaves, NIST 1515 apple leaves, NIST 2709 San Joaquin soil, and NIST 2710 Montana soil) were used to ensure that the instrument calibration was within 5%. Every 10 samples, standards were analyzed to check for instrument drift, and samples of the same material were analyzed in triplicate (coefficient of variance (CV) 8.6±8.0, n=112).

The preserved waters from the leaf washes and water used to water the plants were analyzed for Hg after addition of bromine monochloride (BrCl) and stannous chloride ($SnCl_2$) and subsequent purging of Hg onto gold-coated sand traps. Mercury on traps was quantified using dual amalgamation and cold vapor atomic fluorescence spectrophotometry (CVAFS) (EPA method 1631). The 18.2 MΩ de-ionized water used for leaf washes had an average Hg concentration of 0.2±0.1 ng L^{-1} (n=6).

Methyl Hg concentrations were determined for leaves of *J. scopulorum* and *R. pseudoacacia* collected at the final Harvest (t=17–21 weeks) from this study and from the final Harvest (t=25–29) of the same species that had similar air (5.9±2.3, 30.1±3.5 ng Hg m^{-3}) and soil (0.09±0.02 μg Hg g^{-1}) exposures (Millhollen et al., 2006). Methyl mercury was isolated from foliar tissue by digestion and distillation, then purged onto Tenax traps, and pyrolized to elemental Hg and analyzed with CVAFS detection (Liang, Horvat, & Bloom, 1994). For each set of 12 samples, one water blank and one matrix spike were analyzed. The detection limit ranged from 0.02 to 0.03 ng g^{-1}. Recoveries of matrix spikes were 100±25%. Repeat analysis of the DORM-2 standard (NRCC) had less than 25% error for the daily calibration curves (n=16). Triplicate plant analyses yielded an average CV of 12.7±6.5% (n=21). Reported MeHg concentrations were not blank corrected.

2.3 Foliar mercury fluxes

Foliar Hg fluxes were measured for *J. scopulorum* (n=5,103) beginning 1 month after temperature in the Ecopods was increased, while *R. pseudoacacia* (n=1,620) and *A. tridentata* (n=931) fluxes were measured during the last 2 months of the study (approximately 2.5 months after the temperature in the Ecopods was increased). Foliar Hg fluxes were measured using a cylindrical, polycarbonate (15.2 cm (diameter); 45.7 cm (length)) dynamic field chamber, with a sealed circular sheet of polycarbonate on one end, and a re-sealable plate on the other end that was split into halves with a hole for the plant stem. Ten holes (1.3 cm diameter) were drilled around the circumference along the base of the chamber near the re-sealable end to allow unrestricted airflow into the chamber. A fan was mounted on the permanently sealed end to facilitate air mixing. Mercury concentrations were measured in air entering the chamber adjacent to one of the drilled holes, and exiting the outlet at a port on the sealed end of the chamber using a Hg analyzer (Tekran® 2537A) with a TADS (Tekran® Automated Dual Sampling system) switching unit. Air concentrations were measured every 2.5 min resulting in a flux measurement every 10 min. The flow through the chamber was ~7.0 L min^{-1}, resulting in a 1.2-minute turn over rate. Temperature inside and outside of the chamber (T thermocouples

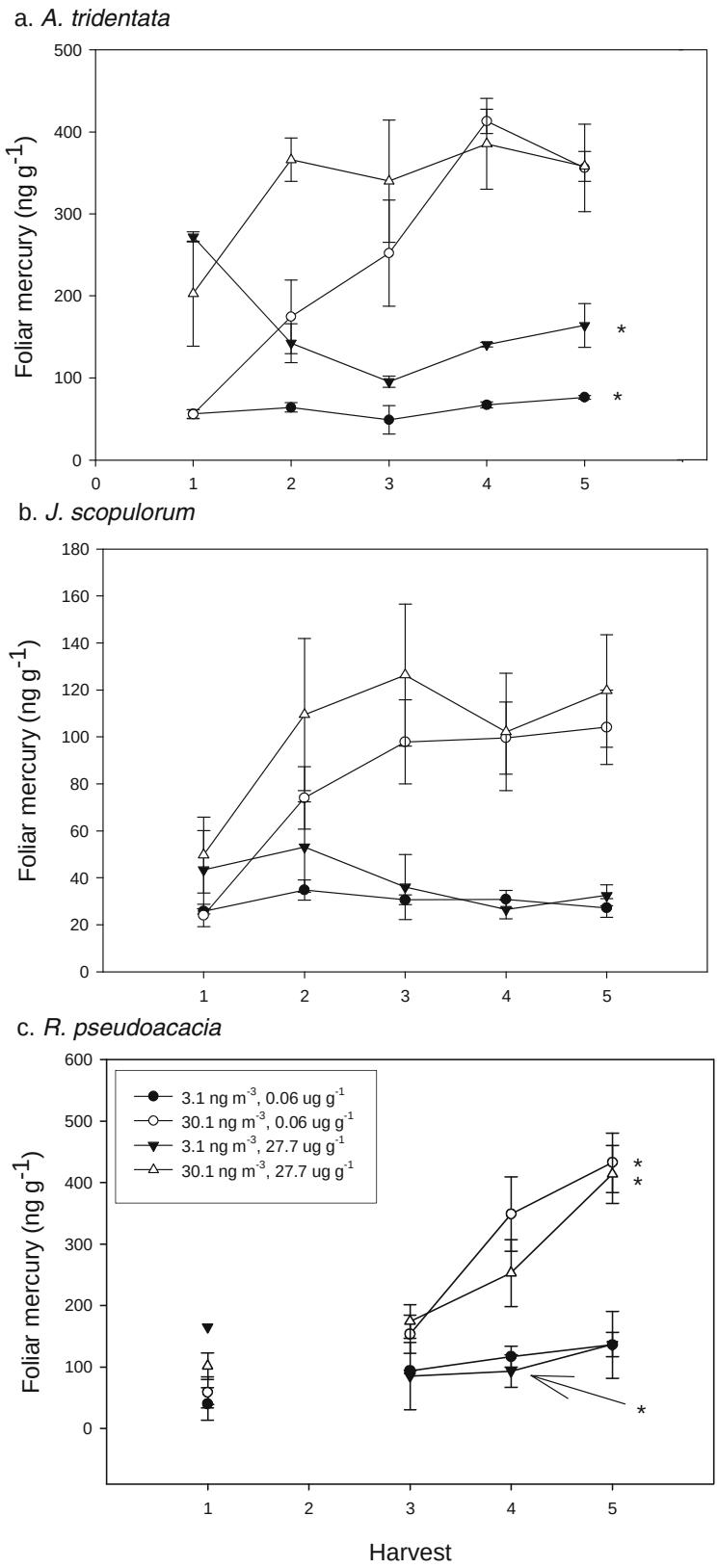

Fig. 2 Foliar mercury concentrations of plant species measured over the course of the experiment as a function of air and soil mercury exposures (explanation in Fig. 2c). Asterisk (*) indicates statistically significant increase in foliar mercury concentration from Harvest 3 to 5 ($p<0.05$)

Omega®) and solar irradiance (LI-COR, Model LI-210), and humidity (Campbell Scientific, Inc., HMP45AC- Temperature and RH probe) were measured and data averaged and recorded every 5 min using a Data logger (Campbell Scientific, Inc., Model 10X).

Foliar Hg flux was determined using the equation:

$$F = ((C_o - C_i)/A)*Q.$$

F is the mercury flux in ng m^{-2} hr^{-1}, C_o and C_i are the concentrations of Hg in the outlet and inlet air in ng m^{-3}, respectively, A is the surface area of the leaf exposed in the chamber in m^2, and Q is the flow rate of air through the chamber in m^3 hr^{-1}. Positive fluxes indicate Hg emission from the foliage and negative fluxes show deposition. Immediately after flux measurements from *R. pseudoacacia* were collected, foliar leaf length was measured and the developed allometric relationship was used to determine foliar exposure area. Leaf area for *J. scopulorum* and *A. tridentata* was determined based on the relationship between this parameter and dry weight following the final Harvest. *J. scopulorum* was harvested 1 month after flux measurements were concluded, this may have resulted in slight underestimation of foliar Hg flux for this plant species grows slowly. The average chamber blank flux was −0.06±0.30 ng m^{-2} hr^{-1} (n=497).

Statistical Analyses were preformed with Minitab® for Windows and a $p<0.05$ was considered significant.

3 Results

There were no statistical differences in leaf wash Hg concentrations as a function of plant species, time, or treatment; therefore these data were lumped for statistical analyses. The average leaf wash concentration was 1.24±0.86 ng m^{-2}. Soil Hg concentrations did not change over time and did not vary with pot depth (data not shown). There were no differences in Hg concentrations of all tissue types (roots, trunk, and foliage) between replicated treatments.

3.1 Foliar mercury

Harvest 1 foliar Hg concentrations, collected immediately after repotting, were in general greater for all plant species in the elevated soil treatments (Fig. 2). The repotted plants were watered and exposed to elevated Hg soil and ambient atmospheric Hg concentrations in the greenhouse for 2 days prior to leaf collection. Prior to Harvest 2, plants grew for 1 month under cold conditions. Harvest 2 foliar Hg concentrations were significantly greater in the elevated air Hg exposures for *A. tridentata* and *J. scopulorum*. The colder temperatures caused *R. pseudoacacia* to senesce.

Higher foliar Hg concentrations in all species were directly correlated with elevated air Hg concentrations ($p<0.01$) (Fig. 2). Foliar Hg concentrations of only *A. tridentata* showed a statistically significant influence of soil Hg concentrations (Fig. 2a, Table 1). Foliar Hg concentrations showed a statistically significant increase from Harvests 3 to 5 for *A. tridentata* in the low air treatments, and for *R. pseudoacacia* in all exposures except ambient air-low soil (Fig. 2c).

3.2 Trunk mercury

Trunk Hg concentrations were higher in the elevated air and elevated soil Hg exposures (Fig. 3) for all plant species suggesting that Hg from both the air and soil influence trunk Hg concentrations (ANOVA calculated F and p values showed a stronger influence of air exposures than soil, Table 1). Mean trunk Hg concentrations for all species in the elevated air and/or soil exposures were (20.9±9.5 ng g^{-1}), and for the ambient air-low soil exposure (8.0±1.5 ng g^{-1}) (Fig. 3).

3.3 Root mercury

J. scopulorum and *R. pseudoacacia* root Hg concentrations were directly correlated with soil Hg concentration ($p<0.005$). There was no difference in root Hg concentrations between species at the low or elevated air Hg concentrations. Roots grown in the elevated soil Hg exposure accounted for >90% of the total plant Hg, with roots grown in the low soil Hg exposure accounting for 20–60% of the total plant Hg (Fig. 4).

3.4 Methyl mercury in foliage

Methyl mercury concentrations were significantly greater for *R. pseudoacacia* foliage than for *J. scopulorum* ($p<0.005$). Although a trend of higher tissue mean MeHg concentration in the elevated air exposure was observed for all plant species, this

Table 1 ANOVA derived (Balanced design) *p* and *F* values, and degrees of freedom (*df*) for comparison of the influence of air and soil mercury exposures on **a** foliar mercury concentrations and **b** trunk mercury concentrations based on the data collected for all three plant species from the final harvest

	J. Scopulorum		*A. tridentata*		*R. pseudoacacia*	
	Air	soil	air	soil	air	soil
a						
p	0.01	0.27	0.01	0.05	0.01	0.26
F	192.1	1.3	133.3	4.71	362.1	1.39
df	39	39	15	15	15	15
b						
p	0.01	0.05	0.05	0.05	0.01	0.5
F	10.29	5.93	4.24	5.92	8.92	0.57
df	19	19	19	19	19	19

A. Foliar concentration statistics.

B. Trunk concentration statistics.

relationship was statistically significant only for two comparisons (Fig. 5). The percentage of MeHg of total leaf Hg was <1% and it was greater for *R. pseudoacacia* (0.72±0.32%) than for *J. scopulorum* (0.19±0.13%).

3.5 Foliar mercury fluxes

Foliar mercury fluxes were not measured for all treatments due to time limitations. In general fluxes were highly variable with no consistent patterns, with deposition and emission of Hg to foliage measured during the day and night (Fay, 2006). However, there were interesting differences between plant species. *J. scopulorum* foliar Hg fluxes were not statistically different from the blank chamber fluxes. In contrast, *R. pseudoacacia* and *A. tridentata* fluxes were more variable than *J. scopulorum* showing net deposition in the elevated air-low soil exposure, and no net exchange or net emission in the ambient air-elevated soil exposure (Fig. 6). The elevated air-low soil exposure showed the greatest variability in foliar Hg fluxes for all treatments and for *R. pseudoacacia* and *A. tridentata* the dominant flux was deposition (Fig. 6). Twenty-four hour average fluxes for *R. pseudoacacia*, at ambient air exposures with both low and elevated soil Hg concentrations, showed no difference between treatments (Fig. 6). A mean flux indicating emission was measured for the ambient air-elevated soil exposure while the flux measured from foliage in the elevated air-low soil exposure exhibited deposition as the dominant flux (Table 2).

4 Discussion

The high mean foliar Hg concentrations measured at Harvest 1, which represented foliage collected in late

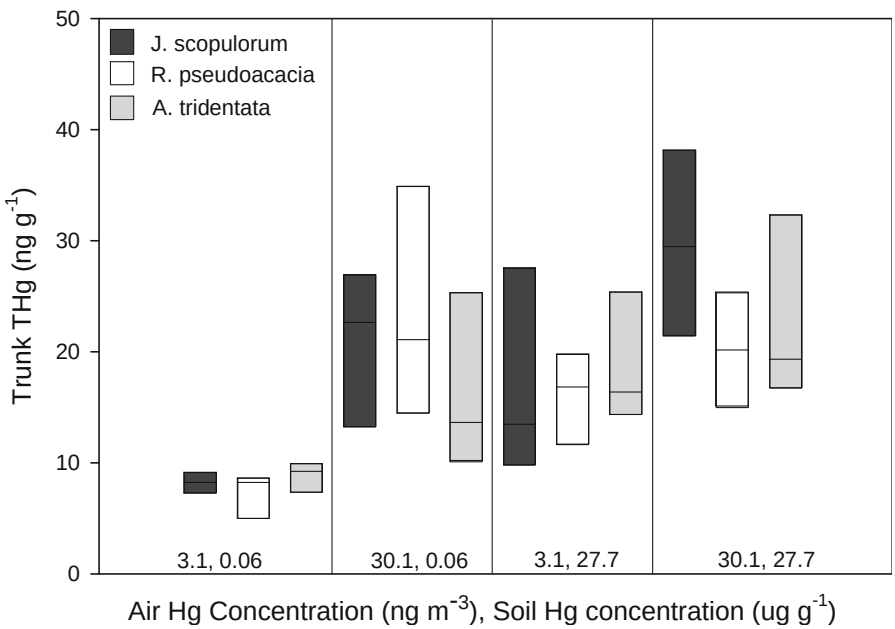

Fig. 3 Trunk mercury concentrations at Harvest 5 (*t*=17–21 weeks) for all three plant species

Table 2 Mean foliar mercury fluxes directly measured with the plant gas exchange chamber

Mercury treatment		Directly measured foliar mercury flux (ng m^{-2} hr^{-1})		
Air (ng m^{-2})	Soil (ug g^{-1})	*A. tridentate* (n=3)	*R. pseudoacacia*	*N*
3.1±0.4	0.06±0.02	–	−0.3±2.9	2
30.1±3.5	27.7±6.3	–	–	
30.13.±5	0.06±0.02	−11.3±18.1	−6.0±9.0	3
3.1±0.4	27.7±6.3	1.4±4.6	−0.2±3.6	5

Negative fluxes indicate deposition and positive flux emission

fall prior to senescence, for all plants in the elevated Hg soil exposures may have been 1) an artifact of repotting where plants may have been exposed to Hg soil dust and would indicate that leaf washing did not remove all particulate bound Hg, 2) due to foliage accumulation of Hg that was volatilized from the soil, or 3) due to root uptake of Hg and translocation to leaf tissue.

The observed consistent increase in foliar Hg concentrations, for *A. tridentata* and *J. scopulorum* from Harvest 1 to 3, which included the cold incubation period, as a function of the high air exposures suggests that evergreen species may accumulate atmospheric Hg year round. At low temperatures (<20°C) evergreen plants are still active but exhibit decreased photosynthesis and respiration (Taiz & Zeiger, 2002). The fact that there was not much increase in concentration for the evergreen species after the third Harvest suggests that the plant tissues of this species may have equilibrated with the high air Hg concentrations. These plant species did not grow rapidly as did the *R. pseudoacacia.* It is noteworthy that the foliar Hg concentration on a per gram basis shown in Fig. 2 does not represent the increase in total Hg concentration of the plant that would occur as plant biomass accumulated over time.

Foliar Hg concentrations increased over time for *R. pseudoacacia* growing in both the high and low Hg soil exposures and for *A. tridentate* at the low soil exposures for the Harvests 3–5 indicating increased accumulation over time. *R. pseudoacacia* foliar tissue accumulated more Hg than *J. scopulorum*, suggesting that deciduous plant species accumulate more Hg than evergreen species.

Soil Hg was found to have a weak influence on *A. tridentata* foliar Hg concentrations (p<0.05, F=4.71, df=15) (Fig. 2, Table 1). Frescholtz et al. (2003) also found a weak soil influence (at soil concentrations ranging from 0.05 to 0.48 μg Hg g^{-1} and air

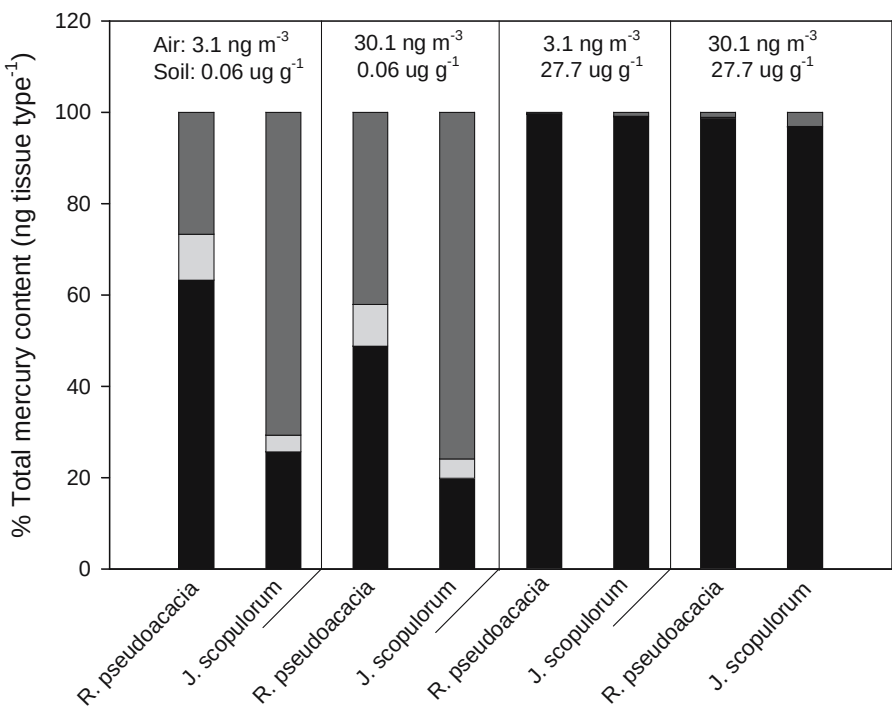

Fig. 4 Percent total mercury for the final harvest of all plant biomass at t=17–21 weeks partitioned into leaves (*dark gray*), trunks (*light gray*), and roots (*black*) as a function of treatment for *R. pseudoacacia* and *J. scopulorum*

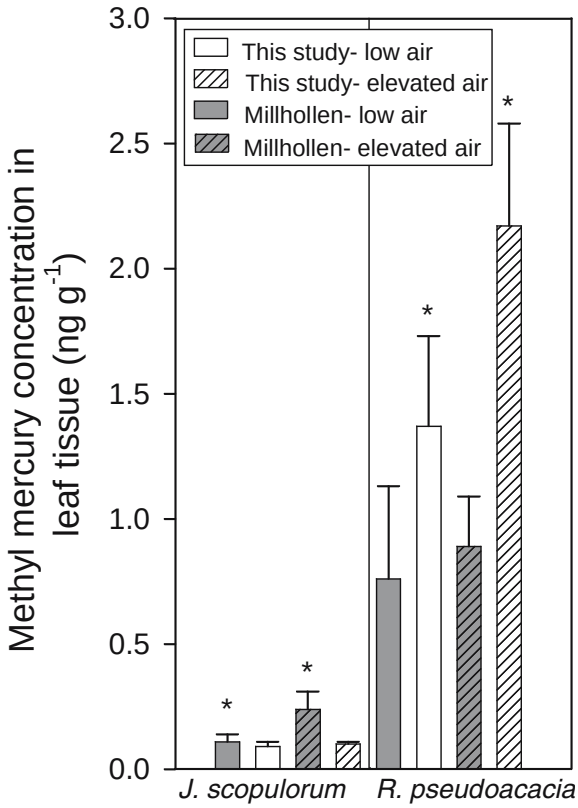

Fig. 5 Methyl mercury concentrations measured in plant tissue collected from this study (t=17–21 weeks) and from the Millhollen et al. (2006) experiment (t=25–29 weeks). Asterisk (*) indicates statistically significant difference between foliar methyl mercury concentrations in the low and elevated air exposures

concentrations ranging from ambient to 70 ng m^{-3}) on foliar Hg concentrations for *P. tremuloides*; while Ericksen et al. (2003) found no soil influence (at soil concentration of 0.03 and 12.3 μg Hg g^{-1} but only one high air exposure concentration). Millhollen et al. (2006) found no soil influence (at soil concentrations of 0.01, 0.09, and 0.9 μg Hg g^{-1} and air exposures similar to this study) for *R. pseudoacacia*, *J. scopulorum*, and *P. ponderosa*. As soil Hg exposure increased, the percentage of Hg allocated to the roots increased (Fig. 4), similar to that reported in other studies (Cocking, Rohrer, Thomas, Walker, & Ward, 1995; Ericksen et al., 2003).

Mercury concentrations of leaf tissue in this study at Harvest 5 (t=17–21 weeks) (Table 3) were in the range of that reported by Millhollen et al. (2006) after an exposure time of 18–23 weeks (Harvests 3 and 4) for *J. scopulorum* (50–130 ng g^{-1}) and *R. pseudoacacia* (150–500 ng g^{-1}). Millhollen et al. (2006) amended soil with $HgCl_2$, whereas this study amended the soil with mine tailing with HgS_2 as the dominant Hg species. Based on this limited data comparison, the Hg species in the soil HgS_2 versus $HgCl_2$, did not have a different effect on foliar Hg concentration.

Trunk Hg concentrations were influenced by both elevated air and soil Hg exposures (Fig. 3, Table 1). Since the pots were covered, isolating the soil, this means that Hg was accumulated from the air directly through the trunks, or transported from the air through

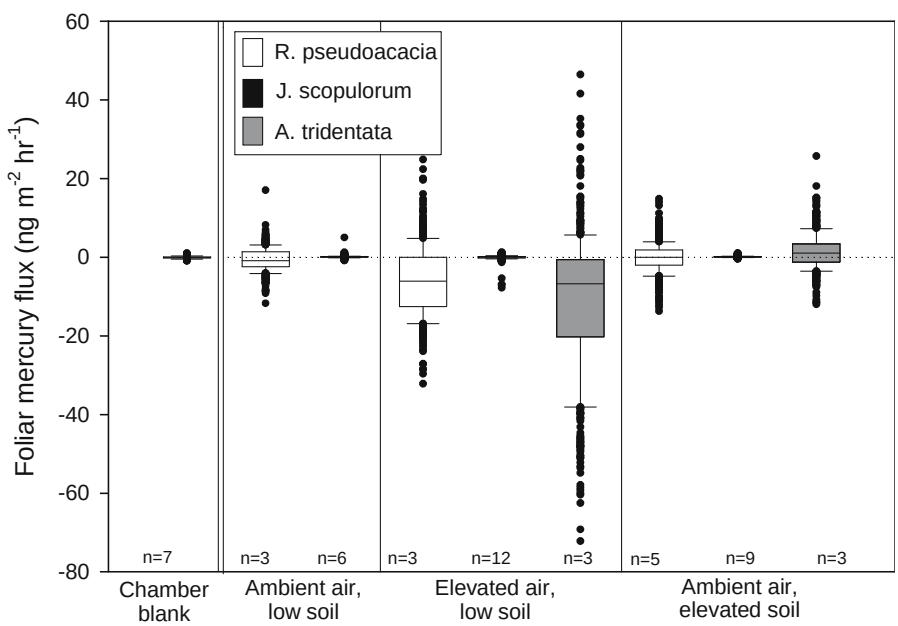

Fig. 6 Foliar mercury fluxes for all three plant species categorized by air and soil mercury exposures. n is equal to the number of days that data was collected; each day represents 24 h of data collection. The chamber blank is given on the left panel of the figure

Table 3 Measured Hg concentrations including: Foliar mercury and methyl mercury concentrations measured in leaf tissue at t=17–21 weeks

Mercury		Foliar total mercury (ng g^{-1})			Foliar methyl mercury (ng g^{-1})	
Air (ng m^{-2})	Soil (ug g^{-1})	*J. scopulorum*	*A. tridentate* (n=3)	*R. pseudoacacia*	*J. scopulorum*	*R. pseudoacacia*
3.1±0.4	0.06±0.02	27.2±3.6	76.4±2.4	136.0±54.2	0.09±0.02	1.37±0.36
30.1±3.5	27.7±6.3	119.6±23.9	357.8±18.3	413.3±46.9	–	–
30.1±3.5	0.06±0.02	104.1±15.9	355.9±53.2	432.1±48.3	–	–
3.1±0.4	27.7±6.3	32.5±4.4	164.1±26.5	136.5±19.7	0.10±0.01	2.17±0.41

stomata to the trunk, and/or originated in the soil and was translocated through the roots to the trunk. The fact that individually elevated air and soil exposures showed a response suggests both influence trunk concentrations. Millhollen et al. (2006) found a similar trend. Frescholtz et al. (2003) measured trunk Hg concentrations in *P. tremuloides* and found a correlation with atmospheric Hg concentrations. Ericksen et al. (2003) hypothesized that trunks can assimilate Hg directly from the air and/or Hg can be translocated from the soil into the trunk. Trunk Hg concentrations from all exposures accounted for ≤10% of the Hg in the entire plant similar to other studies (Ericksen et al., 2003; Millhollen et al., 2006).

Similar to total Hg in foliage, MeHg concentrations and percent MeHg were greater in *R. pseudoacacia*, than in *J. scopulorum* (1 versus 0.3%), and a trend of higher MeHg concentrations as a function of air Hg concentrations was observed. Ericksen et al. (2003) found MeHg concentrations from *P. tremuloides* to be correlated to air Hg concentrations as well.

Table 4 Fluxes calculated based on foliar mercury accumulation using the difference in total mercury concentrations measured between Harvest 3–5 categorized by air and soil mercury exposures

Mercury treatment		Calculated foliar mercury accumulation rates (ng m^{-2} hr^{-1})	
Air (ng m^{-2})	Soil (ug g^{-1})	*A. tridentata*	*R. pseudoacacia*
3.1±0.4	0.06±0.02	–	2.2
30.1±3.5	27.7±6.3	–	–
30.1±3.5	0.06±0.02	5.7	6.0
3.1±0.4	27.7±6.3	3.8	1.8

J. scopulorum fluxes were 0, and not different from blank chamber fluxes. Foliar tissue of this species also showed no significant accumulation of Hg over time. These data suggest that *J. scopulorum* is not very active in Hg cycling.

In contrast, the average 24 h foliar Hg fluxes for *R. pseudoacacia* indicated that deposition of atmospheric Hg to the foliage occurred in all treatments. The highest deposition rates and greatest variability were found in the elevated air-low soil exposure (Fig. 6, Table 2). *A. tridentate* fluxes were similar to *R. pseudoacacia* in all exposures but fluxes were more variable (Fig. 6). *A. tridentate* is an evergreen desert species and the majority of growth occurs in the spring when more water is available (Barker & McKell, 1986). The fact that this species showed greater foliar accumulation and fluxes relative to *J. scopulorum* may be due to the maintained water content of 8–12%, which is high for *A. tridentate*, and allowed for optimal and sustained growth and transpiration. Millhollen et al. (2006) obtained similar results for foliar Hg fluxes for *R. pseudoacacia* (24 hr flux range for all exposures (−6.0 – 6.0 ng m^{-2} hr^{-1}) and for *J. scopulorum* (−2.0 – 2.0 ng m^{-2} hr^{-1}) with more deposition in the elevated air treatment for the former. Ericksen et al. (2003) measured deposition of atmospheric Hg to foliage of aspen at elevated air concentrations.

Mercury accumulation rates as a function of foliar Hg concentrations over time for Harvests 3–5 were calculated only for *R. pseudoacacia* and *A. tridentate* because no statistically significant increase in foliar Hg concentration was found for *J. scopulorum* (Table 4). Foliar Hg accumulation rates were similar to gas exchange chamber measured deposition for *R. pseudoacacia* at the high air Hg exposure (6 ng m^{-2} h^{-1}). However at ambient air exposures, accumulation

rates based on leaf concentrations were an order of magnitude greater than that based on the mean 24 h fluxes. Fluxes measured for *A. tridentata* did not agree with accumulation rates calculated based on foliar Hg concentrations; and Hg emission was measured for plants growing in the ambient air-elevated soil treatment yet Hg was accumulated in this tissue over time. One hypothesis to explain this discrepancy is that flux measured with the plant gas exchange chamber reflects the net flux associated with the foliar surface and the Hg accumulation by the plant is only one component of this net flux.

Average leaf wash mercury concentrations accounted for 0.01–1.52% of the total plant Hg. Since elemental Hg was most likely the dominant form of Hg in the Ecopods (prefiltering of the air could have scrubbed reactive and particulate Hg), the lack of significant accumulation may be due to elemental Hg not being directly deposited to the leaf surface or if deposited to the leaf surface it could have been re-emitted or it was not removed by the water leaf washes.

5 Conclusions

Based on this study and the work of others, foliar tissue concentrations are influenced predominantly by air Hg concentrations, whereas trunk Hg concentrations are influenced by both soil and air Hg exposures, and root Hg concentrations are directly correlated with soil Hg concentrations. Foliar Hg accumulation was greatest in *R. pseudoacacia>A. tridentate>J. scopulorum*. Based on comparison of data collected in this study with that presented by Millhollen et al. (2006), the form of Hg present in the soil (HgS versus $HgCl_2$) had little influence on foliar tissue concentrations and foliar flux. MeHg concentrations were greater in the deciduous species. Since deciduous plant species senesce each year, the leaves will be an annual source of total and MeHg to terrestrial systems. Measured foliar Hg fluxes were bi-directional and variable with the greatest foliar uptake found for the elevated air-low soil Hg treatment and emission of Hg observed for *A. tridentate* in the low air-elevated soil Hg treatment.

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